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# Surface and interface effects in the growth of giant magnetoresistance spin valves for ultrahigh-density data-storage applications

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## Abstract

The current generation of hard-disk drives use giant magnetoresistance (GMR) spin valves as the read-head because the GMR effect is currently the most sensitive way to detect magnetic fields at submicron length scales and data rates of  $\approx 10^8$  Hz. At present, hard-disk capacities are doubling approximately every year. To maintain this growth rate, GMR values will have to double approximately every two years. To achieve such GMR values, major progress will have to be made in controlling surface and interface effects in the growth of GMR spin valves. Published by Elsevier Science Ltd.

*Keywords:* Hard-disk drive; Giant magnetoresistance; Read-heads

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## 1. Introduction

The giant magnetoresistance (GMR) effect is playing a key role in maintaining growth rates in ultrahigh-density data storage in computer hard-disk drives. By the end of the year 2000, all computer hard-disk drives manufactured worldwide will use a GMR thin film as the read-head. At present, GMR films are unequalled in their ability to detect the magnetic fields from bits stored on hard disk at the required data rates of  $\approx 10^8$  Hz.

Fig. 1 illustrates the basic principle of the GMR effect. If two Co films, separated by a Cu film, are magnetized in parallel, majority-spin electrons have a long mean

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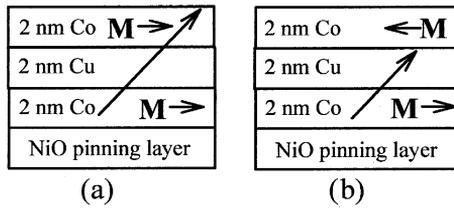


Fig. 1. An illustration of the GMR effect in terms of the change in electron mean free path of majority-spin electrons that occurs when the magnetization of the Co films is switched from parallel to antiparallel. Under the influence of an applied magnetic field the top Co film is free to switch magnetization directions while the bottom Co film has its magnetization pinned in one direction by a layer of NiO.

free path and pass easily through the entire structure. However, if they are magnetized antiparallel, a majority-spin electron from one Co film is a minority-spin electron in the other Co film and its mean free path becomes much shorter. The term “spin valve” was coined to describe this effect in which the magnetization state determines electrical resistivity of the film. In practice, one Co film is magnetically pinned in one direction (often by an antiferromagnetic insulator like NiO) and the other is free to switch orientations easily under the influence of the magnetic field of the magnetic bits stored on the hard disk. Data is read from the hard disk by monitoring the resistance changes as the disk surface passes under the GMR film. The value of the GMR is generally defined as the change in resistance between parallel and antiparallel states divided by the resistance in the parallel state.

The present generation of hard-disk drives has read-heads with GMR values of  $\approx 10\%$  and has storage capacities on the order of 10 Gbytes. However, significantly higher GMR values will be essential if present growth rates in storage density are to be maintained. Fig. 2 illustrates the growth in storage capacity and its relationship to head technology.<sup>1</sup>

Not only has storage capacity grown at an exponential rate, but that exponential rate has been increasing. The famous Moore’s law, according to which microelectronics circuit density doubles every 18 months, corresponds to a growth rate of 60% per year. At present, growth in hard-disk capacity is actually exceeding Moore’s law.

Maintaining this extraordinary growth rate is an issue of great technological and economic importance, and will be one of the key challenges in keeping the information revolution going. For example, millions of hard-disk drives are already used to store the World Wide Web, and the exponential growth commonly expected for the Web will be held back without an exponential growth in storage capacity.

One requirement for storage densities to continue doubling every year (the 100% growth rate in Fig. 2) is that GMR values double approximately every two years. This requirement will be difficult to fulfill. Even today’s best spin valves can scarcely exceed 20% GMR. A variant of GMR technology in which the Cu is replaced with

<sup>1</sup> Fig. 2 was compiled by the first author using numbers taken from advertisements in back issues of popular computer magazines.

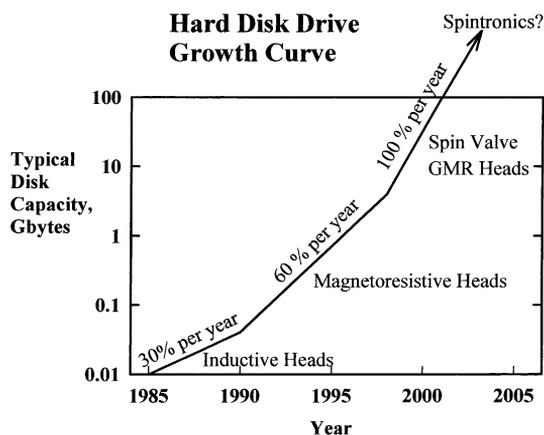


Fig. 2. An illustration of the exponential growth rates in hard-disk drive capacity and the associated read-head technology.

$\text{Al}_2\text{O}_3$  may provide read-heads with values of  $\approx 40\%$ , but even that would provide for only four more years of growth. Beyond that the horizon looks bleak. The emerging field of “Spintronics” might lead to further advances by integrating magnetic and microelectronic devices in some type of spin transistor [1,2], but practical read-heads based on such technology are probably a long way off.

Read-heads are not the only high-density data storage application possible for GMR spin valves. Several companies and research groups are attempting to produce a type of dynamic random access memory chip commonly termed magnetoresistive random access memory (MRAM) [1–3]. Although some working prototypes are being tested, these efforts have had far less impact on memory technology than GMR read-heads. What GMR-MRAM and GMR read-heads have in common is that both technologies benefit greatly from larger GMR values.

Theoretical modeling suggests that an ultimate practical limit for GMR is  $\approx 200\%$  [4,5]. If such a value could be achieved in practice, it would be of great importance to maintaining growth rates. Present growth rates could continue for another eight years. However, the achievement of such values appears highly unlikely without major new efforts to study and improve the surface science aspects of GMR thin film growth. The purpose of this paper is to present a brief account of what would be required.

## 2. Discussion

The theoretical modeling of GMR by Butler et al. [4] is based on a semiclassical Boltzmann transport model including vertex corrections and local fields. In order to estimate the ultimate limit, the model was parameterized to fit the parallel and antiparallel resistivities found in the record-setting GMR values of Parkin in Cu/Co

superlattices [6]. The spin valve consisted of two Co layers separated by a Cu layer. The bulk defect scattering was set to zero, simulating crystalline grains with dimensions considerably exceeding the electron mean free path. The top and bottom surfaces were set to have 100% specular electron scattering, and the Co and Cu layer thicknesses were set to 0.8 nm. The GMR calculated was 207% [5]. Although the GMR value would be still larger if the layers were thinner, the experimental difficulties of achieving thinner films with near-perfect microstructure seems so prohibitive that the results might encourage false hopes.

Fig. 3 identifies some of the key ingredients in achieving the maximum possible GMR in a spin valve. In order to show the largest possible change in electrical resistance, the electrons should have the longest possible electron mean free path when the spin valve is in the parallel magnetization state. One necessary condition is that the top and bottom surfaces of the spin valve scatter electrons specularly. This idea is illustrated in Fig. 4. It should be pointed out that in read-heads the current must flow in the plane of the GMR film so that the film exhibits a measurable resistance. When the current flows perpendicular to the plane, the resistance of the spin valve is so small that it is difficult to measure [7].

Methods have been demonstrated for improving the specularity of GMR spin valves, but none seem to approach 100% specularity. The first evidence of specularity in spin valves was found when NiO was used as the substrate [8]. The data suggested that an atomically sharp metal/insulator interface had some limited tendency to scatter electrons specularly. Transmission electron micrographs suggested that the

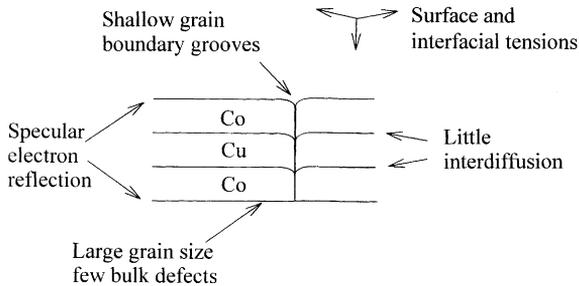


Fig. 3. An illustration of some of the properties that have to be optimized to achieve increased values of the GMR. The inset presents vectors which illustrate the forces acting on a grain boundary groove due to surface and interfacial tensions.

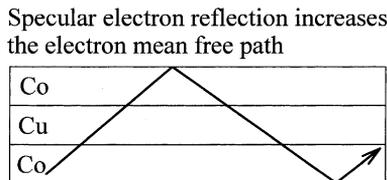


Fig. 4. An illustration of how specular electron reflection at the surfaces can increase the mean free path of electrons in the plane of the film.

first Co layer had a tendency to grow epitaxially on many of the grains in the polycrystalline NiO substrate [9].

Evidence was also found that oxidizing the top of a spin valve could also increase the specularly by producing a metal/insulator interface that was apparently smoother than the as-deposited Co surface [8]. Fig. 5 illustrates why a smoothing might be expected to take place. Protrusions tend to oxidize more readily than a continuous layer of metal atoms so that the surface has a tendency to self-passivate at a sharp metal/insulator interface. Layer-by-layer oxidation is a fairly common occurrence at surfaces [10,11]. Note that the crucial length scale for this smoothness is the Fermi wavelength or about two atomic diameters. Thus, for perfect specularly, an atomically smooth surface or interface is probably essential.

More recent work has confirmed that oxidation can improve the specularly of spin valve surfaces [12,13]. However, the degree of specularly does not appear to exceed about 40% [13]. Clearly, there is an important opportunity here for surface science to find ways to increase the surface specularly.

Also noted in Fig. 3 is the importance of having large grain size and few bulk defects. Grain boundaries and other bulk defects can scatter conduction electrons diffusely, thereby limiting the mean free path. The most obvious way to achieve larger grain size and fewer bulk defects would be to deposit spin valves at elevated temperatures, where the greater mobility of deposited atoms would allow grains to grow larger and defects to anneal out.

Unfortunately, other problems occur at elevated deposition. Fig. 6 illustrates the consequences of deposition at elevated temperature for a NiO\Co\Cu\Co spin valve. The GMR goes to zero only slightly above room temperature. Detailed studies of the failure mechanism suggest that the primary cause is deepening of the grain boundary, which are illustrated in Fig. 3. These grooves are the result of a balance of the forces exerted by surface tension (which acts in the plane of the surface) and interfacial tension (which acts perpendicular to the surface along the grain boundary). These forces are illustrated by the three linked arrows in Fig. 3. At room temperature, these grooves are not at their equilibrium depth (where the force

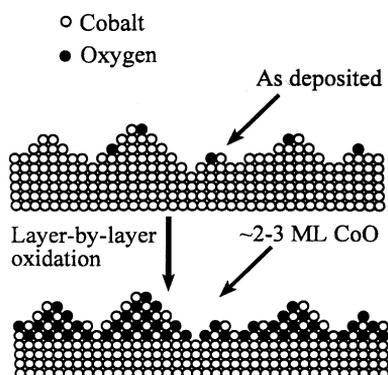


Fig. 5. An illustration of a surface oxidation process that can increase the specular electron scattering at the surface by providing an atomically sharp metal–insulator interface.

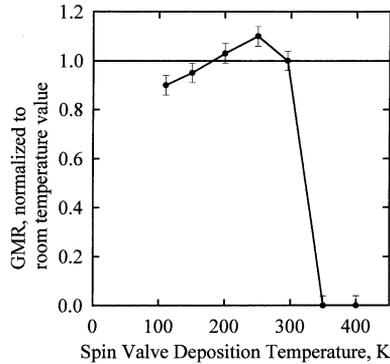


Fig. 6. An illustration of the dependence of the GMR on the substrate temperature during deposition.

vectors would balance, or sum to zero). There is not enough mobility of surface atoms for the depth to reach equilibrium. However, with increasing temperature, the surface mobility increases exponentially. Scanning tunneling microscope studies conducted in vacuum show considerable deepening of the grooves for deposition temperatures as low as 350 K [14].

The problem caused by the grooves is magnetostatic coupling as a result of magnetic poles which are illustrated in Fig. 7. Magnetic poles exist on the surface of protrusions of a magnetic metal. If the roughness is conformal, a magnetic coupling exists between the layers. This coupling is exactly analogous to the attraction force between two bar magnets. If the coupling is too large, in spin valves, it becomes impossible to switch the magnetic state from parallel to antiparallel and no GMR can be observed. This effect is largely responsible for the falloff in GMR in Fig. 6. Note that Fig. 7 uses sinusoidal roughness to illustrate the general phenomenon of the magnetic poles, whereas in actual spin valves the roughness has the morphology illustrated in Fig. 3.

Another factor that must be minimized in spin valves is interdiffusion at the interfaces. Interdiffusion is also promoted by elevated deposition and probably makes a small contribution to the falloff in Fig. 6. It appears that anything that suppresses interdiffusion at the interfaces tends to increase the GMR. One example is the peak in GMR seen in Fig. 6 slightly below room temperature. This effect appears to be due to a lower temperature reducing the amount of interdiffusion that occurs when Co is deposited on Cu [15]. Nevertheless, the GMR drops if the temperature is too low, as

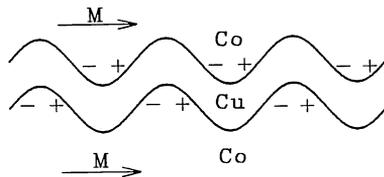


Fig. 7. An illustration of the magnetic poles that exist on roughened magnetic film. These poles lead to a magnetostatic coupling that hinders switching to the antiparallel magnetization state.

shown in Fig. 6. This decrease is due to another effect, the growth of small grains when the entire film is deposited at low temperature [14].

Another effect that can reduce interdiffusion when Co is deposited on Cu is the use of oxygen as a surfactant during spin valve growth [12,16]. Fig. 8 illustrates how oxygen acts as a surfactant. It has been known for some time that oxygen can be a surfactant to suppress intermixing when a high-surface-free-energy metal such as Co is deposited on a low-surface-free-energy metal such as Cu [17,18]. In the absence of a surfactant, Cu tends to segregate onto the surface when Co is deposited, as illustrated in Fig. 8(a). It is well known that Cu atoms are quite mobile on Cu surfaces at room temperature [19]. Gradually the Cu gets left behind in the growing Co film to leave an intermixed region, as illustrated in Fig. 8(b). In the presence of a half monolayer of oxygen adatoms, the situation is completely different. The oxygen atoms bond more strongly to Co than to Cu so that the lowest energy state of the system has the Co atoms at the surface to maximize the number of strong Co–O bonds, as illustrated in Fig. 8(c). Oxygen is quite mobile for room temperature

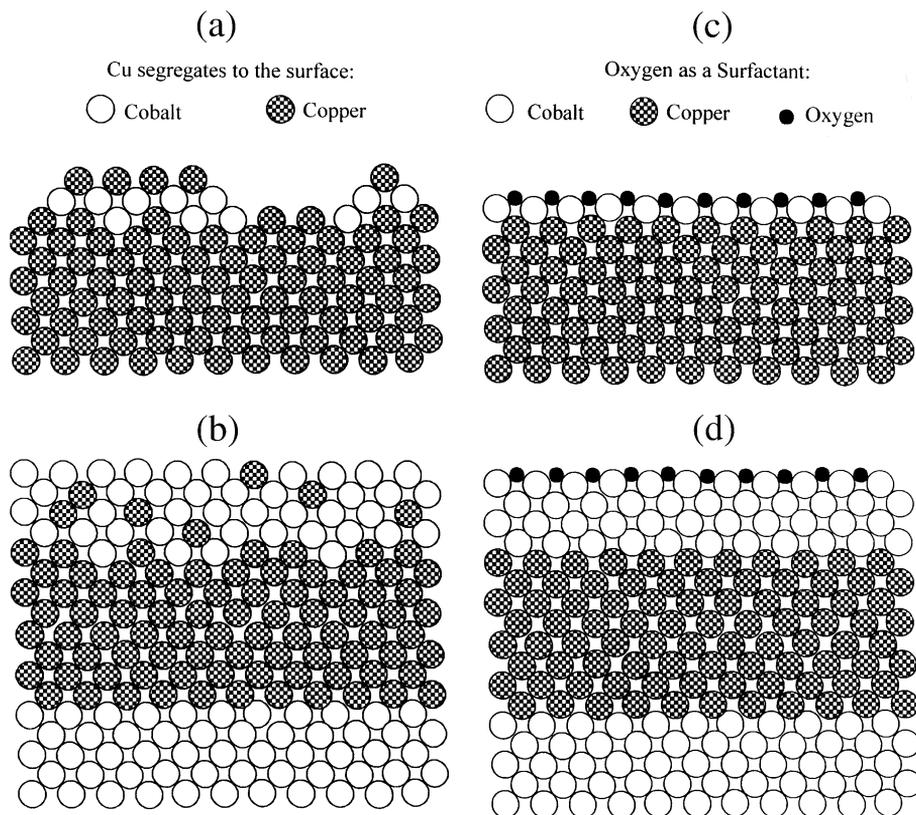


Fig. 8. An illustration in (a) and (b) of how an intermixed layer results from the deposition of Co and Cu under clean conditions and in (c) and (d) of how oxygen may be used as a surfactant to suppress the intermixing.

deposition so it tends to float out to the growing surface leaving behind an interface which is much sharper than without oxygen, as illustrated by comparing Figs. 8(b) and (d). Even with oxygen, however, it is likely that there is a limited amount of intermixing for some crystal planes in these polycrystalline films [20]. All that is known at present is that oxygen appears to reduce the intermixing and increases the GMR in our spin valves typically from 14% to over 16% [12]. It is likely that further increases in GMR could be achieved if there was progress in the surface science of producing perfectly sharp Co/Cu interfaces.

The mechanism for the increase in GMR appears to be a reduction in diffuse scattering of electrons at the interface due to the reduction of intermixing. A perfectly-flat, atomically sharp interface can only transmit or reflect electrons, whereas disorder provides scattering wave vectors (in the sense of Fourier components of the incident electron wave) that can allow diffuse scattering.

Evidence for this effect comes from several observations. First, the resistivity of spin valves is sharply reduced by the use of oxygen as a surfactant [12]. Second, a similar decrease in resistivity and increase in GMR can be achieved by depositing the Co-on-Cu segment of a spin valve at 150 K [15]. Third, deliberate intermixing by co-deposition of Co and Cu increases the resistivity of spin valves and decreases the GMR [21,22]. Thus, all evidence suggests that diffuse scattering of electrons occurs in the intermixed region.

Fig. 9 presents yet another way in which oxygen acts as a surfactant, a smoothing of the surface during growth and that makes electrons scatter more specularly [12].

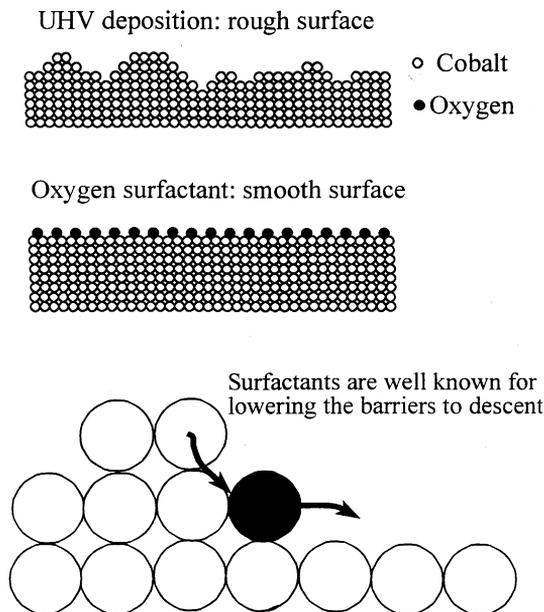


Fig. 9. An illustration of how oxygen acts as a surfactant to smooth a growing surface by lowering the activation-energy barrier for descent of an upper terrace atom to a lower terrace.

The explanation for this effect is probably that oxygen acts to lower the so-called Schwoebel activation-energy barrier for metal atoms to descend to a lower terrace. Because of the random arrival of deposited atoms, some roughness of the surface is expected. To smooth out this roughness, upper-terrace atoms need to overcome the Schwoebel barrier. It is well established in research on epitaxial growth of single crystals that just a mechanism occurs; in this mechanism a surfactant gives a more layer-by-layer growth mode and thus a flatter surface [23–32]. Apparently, the barrier is lowered by a process involving the concerted motion of the metal and surfactant atoms so that the effective number of chemical bonds that must be broken in getting over the barrier is reduced. Of course, it is not likely that our efforts along these lines have produced perfect specular scattering, and there is room for improvement.

In overview, it seems clear that much larger GMR values could be obtained in spin valves if increased control over film growth were possible. However, the challenges are great. For example, in order to make grain size far larger than the  $\approx 10$  nm typical of spin valves high deposition temperatures seem unavoidable. At such temperatures surfactants seem to work poorly. It is possible that with enough research breakthroughs in finding new surfactants could occur. It is also possible that unforeseen breakthroughs in achieving greater control over film growth will occur. However, one thing seems certain. Without a great deal more research on the surface science of thin film growth in general and of GMR films in particular, it is unlikely that GMR values will ever reach their potential.

### 3. Conclusions

The major conclusions of this paper may be summarized as follows:

1. In order to continue exponential growth rates in computer hard-disk storage capacity, major progress will have to be made in magnetoresistance values of the read-head.
2. If no progress is made, hard-disk capacity will begin to suffer within two years and become a critical problem within four years.
3. The theoretical limit to the GMR in spin valves is about an order of magnitude larger than can presently be achieved.
4. If this theoretical limit could be achieved in practice, it would provide GMR read-heads of adequate sensitivity to maintain current growth rates for about another eight years.
5. A major increase in research on surface science aspects of GMR film growth is needed if substantial increases in GMR are to be achieved.

### References

- [1] S.A. Wolf, *J. Supercond.* 13 (2000) 195.
- [2] G.A. Prinz, *J. Mag. Mater.* 200 (1999) 57.

- [3] K. Bussmann, S.F. Cheng, G.A. Prinz, Y. Hu, R. Gutmann, D. Wang, R. Beech, J. Zhu, *IEEE Trans. Mag.* 34 (1998) 924.
- [4] W.H. Butler, X.-G. Zhang, J.M. MacLaren, *J. Supercond.* 13 (2000) 221.
- [5] W.H. Butler, to be published.
- [6] S.S.P. Parkin, to be published.
- [7] W.P. Pratt Jr., S.-F. Lee, J.M. Slaughter, R. Loloe, P.A. Schroeder, J. Bass, *Phys. Rev. Lett.* 66 (1991) 3060.
- [8] W.F. Egelhoff Jr., T. Ha, R.D.K. Misra, Y. Kadmon, J. Nir, C.J. Powell, M.D. Stiles, R.D. McMichael, C.-L. Lin, J.M. Sivertsen, J.H. Judy, K. Takano, A.E. Berkowitz, T.C. Anthony, J.A. Brug, *J. Appl. Phys.* 78 (1995) 273.
- [9] H.D. Chopra, B.J. Hockey, P.J. Chen, W.F. Egelhoff Jr., M. Wuttig, S.Z. Hua, *Phys. Rev. B* 55 (1997) 8390.
- [10] N. Niyata, H. Watanabe, M. Ichikawa, *Appl. Phys. Lett.* 72 (1998) 1715.
- [11] M. Yata, H. Rouch, *Appl. Phys. Lett.* 75 (1999) 1021.
- [12] W.F. Egelhoff Jr., P.J. Chen, C.J. Powell, M.D. Stiles, R.D. McMichael, J.H. Judy, K. Takano, A.E. Berkowitz, *J. Appl. Phys.* 82 (1998) 6142.
- [13] S.X. Wang, K. Yamada, W.E. Bailey, *IEEE Trans. Mag.*, in press.
- [14] D.C. Parks, P.J. Chen, W.F. Egelhoff Jr., R.D. Gomez, *J. Appl. Phys.* 87 (2000) 3023.
- [15] W.F. Egelhoff Jr., T. Ha, R.D.K. Misra, C.J. Powell, M.D. Stiles, R.D. McMichael, C.-L. Lin, J.M. Sivertsen, J.H. Judy, *J. Appl. Phys.* 79 (1996) 282.
- [16] C. Tölkes, R. Struck, R. David, P. Zeppenfeld, G. Comsa, *Appl. Phys. Lett.* 73 (1998) 1059.
- [17] D.A. Steigerwald, I. Jacob, W.F. Egelhoff Jr., *Surf. Sci.* 202 (1988) 472.
- [18] W.F. Egelhoff Jr., D.A. Steigerwald, *J. Vac. Sci. Technol. A7* (1989) 2167.
- [19] C. Klünker, J.B. Hannon, M. Giesen, H. Ibach, G. Boisvert, L.J. Lewis, *Phys. Rev. B* 58 (1998) R7556.
- [20] W.L. Ling, O. Takeuchi, D.F. Ogletree, Z.Q. Qiu, M. Salmeron, *Surf. Sci.* 450 (2000) 227.
- [21] M. Suzuki, Y. Taga, *J. Appl. Phys.* 74 (1993) 4660.
- [22] W.F. Egelhoff Jr., C.J. Powell, R.D. McMichael, P.J. Chen, unpublished results.
- [23] R. Kunkel, B. Poelsema, L.K. Verheij, G. Comsa, *Phys. Rev. Lett.* 65 (1990) 733.
- [24] J. Jacobsen, K.W. Jacobsen, P. Stoltze, J.K. Nørskov, *Phys. Rev. Lett.* 74 (1995) 2295.
- [25] H.A. van der Vegt, H.M. van Pinxteren, M. Lohmeier, E. Vlieg, J.M.C. Thornton, *Phys. Rev. Lett.* 68 (1992) 3335.
- [26] B. Poelsema, R. Kunkel, N. Nagel, A.F. Becker, G. Rosenfeld, G. Comsa, *Appl. Phys. A* 53 (1991) 369.
- [27] S. Esch, M. Hohage, T. Michely, G. Comsa, *Phys. Rev. Lett.* 72 (1994) 518.
- [28] C.W. Oh, E. Kim, Y.H. Lee, *Phys. Rev. Lett.* 76 (1996) 776.
- [29] H.A. van der Vegt, J. Alvarez, X. Torrelles, S. Ferrer, E. Vlieg, *Phys. Rev. B* 52 (1995) 17443.
- [30] H.A. van der Vegt, M. Breeman, S. Ferrer, V.H. Etgens, S. Ferrer, V.H. Etgens, X. Torrelles, P. Fajardo, E. Vlieg, *Phys. Rev. B* 51 (1995) 14806.
- [31] J. Vrijmoeth, H.A. van der Vegt, J.A. Meyer, E. Vlieg, R.J. Behm, *Phys. Rev. Lett.* 72 (1994) 3843.
- [32] Z. Zhang, M.G. Lagally, *Phys. Rev. Lett.* 72 (1994) 693.